

TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371

206228US0PCT

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR

09/807425

INTERNATIONAL APPLICATION NO.
PCT/IP99/05765INTERNATIONAL FILING DATE
19 OCTOBER 1999PRIORITY DATE CLAIMED
20 OCTOBER 1998 (earliest)

TITLE OF INVENTION

LIQUID CRYSTAL OPTICAL ELEMENT AND METHOD FOR ITS PRODUCTION

APPLICANT(S) FOR DO/EO/US

Satoshi NIYAMA, et al.


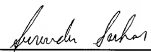
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (24) indicated below.
4. ☒ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371 (c) (2))
 - a. ☐ is attached hereto (required only if not communicated by the International Bureau).
 - b. ☒ has been communicated by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
 - a. ☒ is attached hereto.
 - b. ☐ has been previously submitted under 35 U.S.C. 154(d)(4).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3))
 - a. ☐ are attached hereto (required only if not communicated by the International Bureau).
 - b. ☐ have been communicated by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☒ have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☐ An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).
10. ☐ An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).
11. ☐ A copy of the International Preliminary Examination Report (PCT/IPEA/409).
12. ☒ A copy of the International Search Report (PCT/ISA/210).

Items 13 to 20 below concern document(s) or information included:

13. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
14. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
15. ☐ A **FIRST** preliminary amendment.
16. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
17. ☐ A substitute specification.
18. ☐ A change of power of attorney and/or address letter.
19. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
20. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
21. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
22. ☐ Certificate of Mailing by Express Mail
23. ☒ Other items or information:

Request for Consideration of Documents Cited in International Search Report
Notice of Priority / PCT/IB/304 / Drawings (1 sheet) / PCT/IB/308

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR) <div style="font-size: 24pt; font-weight: bold; margin-top: 5px;">09/807425</div>		INTERNATIONAL APPLICATION NO. <div style="font-weight: bold; margin-top: 5px;">PCT/JP99/05765</div>		ATTORNEY'S DOCKET NUMBER <div style="font-weight: bold; margin-top: 5px;">206228US0PCT</div>																																									
24. The following fees are submitted: BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)) : <input type="checkbox"/> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO \$1000.00 <input checked="" type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO \$860.00 <input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$710.00 <input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$690.00 <input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) \$100.00				CALCULATIONS PTO USE ONLY <table border="1" style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 80%;">ENTER APPROPRIATE BASIC FEE AMOUNT =</td> <td style="width: 20%; text-align: right; font-weight: bold;">\$860.00</td> </tr> <tr> <td>Surcharge of \$130.00 for furnishing the oath or declaration later than months from the earliest claimed priority date (37 CFR 1.492 (e)). <input type="checkbox"/> 20 <input checked="" type="checkbox"/> 30</td> <td style="text-align: right; font-weight: bold;">\$130.00</td> </tr> <tr> <td> <table border="1" style="width: 100%; border-collapse: collapse;"> <tr> <th style="width: 25%;">CLAIMS</th> <th style="width: 25%;">NUMBER FILED</th> <th style="width: 25%;">NUMBER EXTRA</th> <th style="width: 25%;">RATE</th> </tr> <tr> <td>Total claims</td> <td style="text-align: center;">- 20 =</td> <td style="text-align: center;">0</td> <td>x \$18.00</td> </tr> <tr> <td>Independent claims</td> <td style="text-align: center;">- 3 =</td> <td style="text-align: center;">0</td> <td>x \$80.00</td> </tr> </table> </td> <td style="text-align: right; font-weight: bold;">\$0.00</td> </tr> <tr> <td>Multiple Dependent Claims (check if applicable). <input type="checkbox"/></td> <td style="text-align: right; font-weight: bold;">\$0.00</td> </tr> <tr> <td>TOTAL OF ABOVE CALCULATIONS =</td> <td style="text-align: right; font-weight: bold;">\$990.00</td> </tr> <tr> <td><input checked="" type="checkbox"/> Applicant claims small entity status. (See 37 CFR 1.27). The fees indicated above are reduced by 1/2.</td> <td style="text-align: right; font-weight: bold;">\$0.00</td> </tr> <tr> <td>SUBTOTAL =</td> <td style="text-align: right; font-weight: bold;">\$990.00</td> </tr> <tr> <td>Processing fee of \$130.00 for furnishing the English translation later than months from the earliest claimed priority date (37 CFR 1.492 (f)). <input type="checkbox"/> 20 <input type="checkbox"/> 30 +</td> <td style="text-align: right; font-weight: bold;">\$0.00</td> </tr> <tr> <td>TOTAL NATIONAL FEE =</td> <td style="text-align: right; font-weight: bold;">\$990.00</td> </tr> <tr> <td>Fees for recording the enclosed assignment (37 CFR 1.21(h)). 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a. <input checked="" type="checkbox"/> A check in the amount of <u>\$990.00</u> to cover the above fees is enclosed. b. <input type="checkbox"/> Please charge my Deposit Account No. _____ in the amount of _____ to cover the above fees. A duplicate copy of this sheet is enclosed. c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>15-0030</u> . A duplicate copy of this sheet is enclosed. d. <input type="checkbox"/> Fees are to be charged to a credit card. WARNING: Information on this form may become public. Credit card information should not be included on this form. Provide credit card information and authorization on PTO-2038.																																													
NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.																																													
SEND ALL CORRESPONDENCE TO: <div style="border: 1px solid black; padding: 10px; margin-top: 10px; width: 40%;">  <div style="font-size: 24pt; font-weight: bold; margin-top: 5px;">22850</div> <div style="text-align: center; margin-top: 10px;"> Surinder Sachar Registration No. 34,423 </div> </div> <div style="margin-top: 10px;"> <div style="text-align: center;">  SIGNATURE Norman F. Oblon NAME <div style="font-weight: bold; margin-top: 5px;">24,618</div> REGISTRATION NUMBER <div style="text-align: center; margin-top: 10px;"> <u>April 20 2001</u> DATE </div> </div> </div>																																													

206228US-0 PCT

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF: :
SATOSHI NIIYAMA ET AL : ATTN: APPLICATION DIVISION
SERIAL NO: 09/807,425
FILED: April 20, 2001 :
FOR: LIQUID CRYSTAL OPTICAL ELEMENT
AND METHOD FOR ITS PRODUCTION

PRELIMINARY AMENDMENT

ASSISTANT COMMISSIONER FOR PATENTS
WASHINGTON, D.C. 20231

SIR:

Prior to examination on the merits, please amend the above-identified application as follows.

IN THE CLAIMS

Please amend the claims as shown on the marked-up copy following this amendment to read as follows.

3. (Amended) The method for producing a liquid crystal optical element according to Claim 1, wherein each of R_1 and R_2 which are independent of each other, is an ethylene group or a propylene group.

4. (Amended) The method for producing a liquid crystal optical element according to Claim 1, wherein each of A_1 and A_2 which are independent of each other, is an acryloyl group or a methacryloyl group.

5. (Amended) The method for producing a liquid crystal optical element according to Claim 1, wherein each of n and m which are independent of each other, is from 1 to 4.

8. (Amended) The method for producing a liquid crystal optical element according to Claim 6, wherein the two types of curable compounds have curable sites connectable to each other.

9. (Amended) The method for producing a liquid crystal optical element according to Claim 6, which contains a curable compound having a molecular weight of at least 1,000.

10. (Amended) The method for producing a liquid crystal optical element according to Claim 1, wherein the mixture contains a chiral agent.

11. (Amended) The method for producing a liquid crystal optical element according to Claim 1, wherein the mixture contains a chiral agent, and the helical pitch of the chiral agent is at least 4 μm and at most three times of the electrode gap.

13. (Amended) The method for producing a liquid crystal optical element according to Claim 11, wherein the helical pitch is at least 5 μm and at most two times of the electrode gap.

14. (Amended) The method for producing a liquid crystal optical element according to Claim 1, wherein the mixture contains a very small amount of a curing catalyst.

15. (Amended) The method for producing a liquid crystal optical element according to Claim 1, wherein a plurality of compounds of the formula (1) wherein n and m are different, are used in combination.

16. (Amended) A liquid crystal optical element produced by the method as defined in Claim 1.

17. (New) The method for producing a liquid crystal optical element according to Claim 6, wherein the mixture contains a chiral agent.

18. (Amended) The method for producing a liquid crystal optical element according to Claim 6, wherein the mixture contains a chiral agent, and the helical pitch of the chiral agent is at least 4 μm and at most three times of the electrode gap.

19. (New) The method for producing a liquid crystal optical element according to Claim 18, wherein the electrode gap is from 4 to 50 μm .

20. (New) The method for producing a liquid crystal optical element according to Claim 18, wherein the helical pitch is at least 5 μm and at most two times of the electrode gap.

21. (New) The method for producing a liquid crystal optical element according to Claim 18, wherein the mixture contains a very small amount of a curing catalyst.

22. (New) The method for producing a liquid crystal optical element according to Claim 18, wherein a plurality of compounds of the formula (1) wherein n and m are different, are used in combination.

23. (New) A liquid crystal optical element produced by the method as defined in Claim 18.

REMARKS

Claims 1-23 are active in the present application. Claims 3-5, 8-11 and 13-16 have been amended to remove multiple dependencies. Support for new Claims 17-23 is found in Claims 10-16. No new matter is added. An action on the merits and allowance of claims is solicited.

Respectfully submitted,

OBLON, SPIVAK, McCLELLAND,
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Serial No: 09/807,425
Amendment Filed on:

--3. (Amended) The method for producing a liquid crystal optical element according to Claim 1 [or 2], wherein each of R_1 and R_2 which are independent of each other, is an ethylene group or a propylene group.

4. (Amended) The method for producing a liquid crystal optical element according to Claim 1[, 2 or 3], wherein each of A_1 and A_2 which are independent of each other, is an acryloyl group or a methacryloyl group.

5. (Amended) The method for producing a liquid crystal optical element according to Claim 1[, 2, 3 or 4], wherein each of n and m which are independent of each other, is from 1 to 4.

8. (Amended) The method for producing a liquid crystal optical element according to Claim 6 [or 7], wherein the two types of curable compounds have curable sites connectable to each other.

9. (Amended) The method for producing a liquid crystal optical element according to Claim 6[, 7 or 8], which contains a curable compound having a molecular weight of at least 1,000.

10. (Amended) The method for producing a liquid crystal optical element according to [any one of Claims 1 to 9] Claim 1, wherein the mixture contains a chiral agent.

11. (Amended) The method for producing a liquid crystal optical element according to [any one of Claims 1 to 9] Claim 1, wherein the mixture contains a chiral agent, and the helical pitch of the chiral agent is at least 4 μm and at most three times of the electrode gap.

13. (Amended) The method for producing a liquid crystal optical element according to Claim 11 [or 12], wherein the helical pitch is at least 5 μm and at most two times of the electrode gap.

14. (Amended) The method for producing a liquid crystal optical element according to [any one of Claims 1 to 13] Claim 1, wherein the mixture contains a very small amount of a curing catalyst.

15. (Amended) The method for producing a liquid crystal optical element according to [any one of Claims 1 to 14] Claim 1, wherein a plurality of compounds of the formula (1) wherein n and m are different, are used in combination.

16. (Amended) A liquid crystal optical element produced by the method as defined in [any one of Claims 1 to 15] Claim 1.--

Claims 17-23 (New).

DESCRIPTIONLIQUID CRYSTAL OPTICAL ELEMENT AND METHOD FOR ITS
PRODUCTIONTECHNICAL FIELD

- 5 The present invention relates to a liquid crystal optical element whereby the transmittance, scattering and reflection state of the element is controlled by application/non-application of an electric field and which is useful for e.g. a light-controlling element, a display element or an optical shutter.
- 10

BACKGROUND ART

- A transmission/scattering type optical element has been proposed in which a liquid crystal and a transparent polymer are combined to form a difference in the refractive index between the polymer and the liquid crystal or in the interior of the liquid crystal (among microscopic regions). It is called, for example, a liquid crystal/polymer composite element, a liquid crystal/resin composite element or a dispersion type liquid crystal element. In principle, this element requires no polarizing plate, whereby the light absorption loss is small, and a high scattering performance can be obtained, and thus, it has a significant merit in that the light utilizing efficiency in the entire element is high.
- 15
- 20
- 25

By utilizing such characteristics, it is employed for a light-controlling glass, an optical shutter, a

laser apparatus and a display apparatus. One showing a scattering state under application of no voltage and a transparent state under application of a voltage, has been commercialized.

5 Further, prior art reference 1 (USP 5,188,760) discloses an element employing a liquid crystal and a polymerizable liquid crystal. According to this prior art reference 1, under application of no voltage, the element shows a transparent state as observed from any
10 direction as the liquid crystal and the polymerized liquid crystals in the element have the same alignment direction, and under application of a voltage, the alignment of the liquid crystals in the element is controlled by an electric field, and the alignment
15 direction of liquid crystal molecules changes variously in the microscopic regions, whereby the element shows a scattering state.

Further, it has been disclosed that the contrast ratio can be improved by adding a chiral agent to provide
20 a helical structure in the initial alignment. This element is called "an anisotropic gel" or "a liquid crystal gel". In this prior art reference 1, a mesogen monomer having acryloyl groups at the terminals was used.

Further, prior art reference 2 (PCT International
25 Publication WO92/19695) also discloses an element having a similar structure. The operation mode was the same as in prior art reference 1, and a very small amount of a

polymer is dispersed in the chiral nematic liquid crystal to obtain a transparent state under application of no voltage and a scattering state under application of a voltage. This element is called PSCT (polymer stabilized
5 cholesteric texture). Also in this prior art reference 2, a mesogen monomer having acryloyl groups at the terminals, was disclosed.

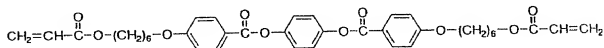
Characteristics of a liquid crystal optical device obtained by preparing a mixture comprising a liquid
10 crystal and an uncured curable compound, and curing the curable compound to form a liquid crystal/cured composite layer, depend largely on the structure of the liquid crystal/cured composite. Further, the molecular structure of the uncured curable compound to be used,
15 will give a substantial influence over the structure of the formed liquid crystal/cured composite.

It has been reported that in general, with a curable compound containing a mesogen structure such as a biphenyl structure, curable sites at both ends will bond,
20 the modulus of elasticity after being cured is large, and the glass transition temperature of the obtainable polymer is also high.

On the other hand, this means that restrictions are given to the free volume and the molecular motion of the
25 curable compound during the curing, and at the later stage of the curing process, the reactivity of the curable sites is likely to be suppressed, and there has

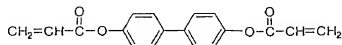
been a problem that the curing reaction tends to be not enough, or a very long curing time will be required.

Namely, as the structures of uncured curable compounds, prior art reference 1 has disclosed the compound of the formula (2), and the prior art reference 2 has disclosed the compound of the formula (3);



Formula (2)

10



Formula (3)

However, the cured products formed by using these compounds alone had characteristics attributable to the molecular structures. Namely, with the element disclosed in prior art reference 1, it is essential to use a monomer having a liquid crystal nature, and accordingly, the uncured curable compound contains a highly crystallizable rigid mesogen structural portion as shown in the formula (2), while with the compound of the formula (3) to be used in prior art reference (2), the distance between the mesogen structural portion and the curable site (an acryloyl group in this case) is short, and accordingly, the molecular weight between the crosslinking points tends to be small, whereby the obtained cured products tend to be hard and brittle.

Further, for the same reason, the mobility of the uncured sites during the curing will be substantially impaired, whereby there has been a problem that a long curing time is required for adequate curing.

5 Further, with the liquid crystal optical elements of the prior art references, the voltage transmittance curves of the elements were likely to change by driving of application of an electric field for a plurality of times, and the contrast between application and non-
10 application of an electric field, was still low. Particularly, the physical properties of the resin as the cured product of the liquid crystal/cured composite layer, related to the electro-optical characteristics of the liquid crystal optical element, and if the modulus of
15 elasticity of the resin was too high or too brittle, the required driving voltage tended to be high, and at a relatively low driving voltage range, no adequate contrast ratio in the reflectance change or in the transmittance change was sometimes obtained between
20 application and non-application of a voltage.

Further, as a means to improve the contrast ratio in the transmittance (or the reflectance) of a liquid crystal optical element obtainable between application and non-application of a voltage in the prior art, prior
25 art reference 1 proposes to add a chiral agent to the mixture and to introduce a helical structure to the alignment mode of the curable compound after the curing,

and prior art reference 2 proposes to add a chiral agent to bring the helical pitch to a level of from 0.5 to 4 μm .

However, the addition of the chiral agent may sometimes cause a problem such that it increases the driving voltage of the element or it decreases the transmittance when the element is transparent. Further, when a mixture of a liquid crystal with an uncured curable compound, is injected into a liquid crystal cell, or when it is sandwiched between substrates provided with transparent electrodes, such as resin films provided with electrodes, if the chiral agent is contained in a large amount, there will be a problem that injection irregularities or sandwiching irregularities are likely to result.

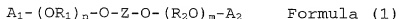
The present invention provides a liquid crystal optical element having high reliability and high contrast, whereby the voltage transmittance curve of the element will not substantially change even by driving of application or non-application of an electric field for a plurality of times. Further, it provides a production method whereby a liquid crystal optical element can be produced easily and constantly in good yield.

Further, it provides a liquid crystal optical element which can be produced, for example, in a short period of curing time and which has a high contrast ratio even at a low driving voltage.

Further, it provides a liquid crystal optical element, whereby addition of the chiral agent can be minimized, and a high contrast ratio can be obtained in the transmittance characteristics obtainable at the time of application and non-application of a voltage, and the driving voltage may be not increased as far as possible.

DISCLOSURE OF THE INVENTION

The present invention provides a method for producing a liquid crystal optical element, which comprises sandwiching a mixture of a liquid crystal with an uncured curable compound between a pair of substrates which are provided with transparent electrodes and of which at least one is transparent, and curing the curable compound to form a liquid crystal/cured composite layer, wherein the curable compound contains a compound of the formula (1):



wherein each of A_1 and A_2 which are independent of each other, is an acryloyl group, a methacryloyl group, a glycidyl group or an allyl group; each of R_1 and R_2 which are independent of each other, is a C_{2-6} alkylene group; Z is a bivalent mesogen structure; and each of n and m which are independent of each other, is an integer of from 1 to 10.

Further, in one embodiment, the present invention provides the above-mentioned method wherein Z is a 4,4'-biphenylene group, or a 4,4'-biphenylene group having

part or all of hydrogen substituted by C_{1-2} alkyl or halogen atoms.

Further, in one embodiment, the present invention provides the production method wherein each of R_1 and R_2 which are independent of each other, is an ethylene group or a propylene group.

Further, in one embodiment, the present invention provides the production method wherein each of A_1 and A_2 which are independent of each other, is an acryloyl group or a methacryloyl group.

Further, in one embodiment, the present invention provides the production method wherein each of n and m which are independent of each other, is from 1 to 4.

Further, in one embodiment, the present invention provides the production method wherein the curable compound contains two types of curable compounds, of which the molecular weights are different by at least two times. In the above production method, three or more compounds having different molecular weights may be employed, so long as at least two types of compounds among them satisfy the above-mentioned conditions.

Further, in one embodiment, the present invention provides the method for producing a liquid crystal optical device wherein the curable compound contains a curable compound containing a mesogen structural portion in its molecule and a curable compound containing no mesogen structural portion.

Further, in one embodiment, the present invention provides the method for producing a liquid crystal optical element wherein the mixture contains a chiral agent, and the helical pitch of the chiral agent is at least 4 μm and at most three times of the electrode gap. Thus, by employing a large helical pitch which has not heretofore been employed, a high contrast ratio and a low driving voltage can be satisfied at the same time.

Further, the present invention provides a liquid crystal optical element produced by means of the above production method.

BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 is a flow chart showing an embodiment of the method for producing a liquid crystal optical element of the present invention.

Figure 2 is a diagrammatic cross-sectional view of an embodiment of the liquid crystal optical element of the present invention.

Figure 3 is a diagrammatic view showing an embodiment of use of the liquid crystal optical element of the present invention.

BEST MODE FOR CARRYING OUT THE INVENTION

In the present invention, an oxyalkylene structure having a high molecular mobility is introduced between the mesogen structural portion and the curable site in the uncured curable compound, whereby the molecular mobility at the curable site in the curing process can be

improved, and it is possible to obtain a liquid crystal optical element which has a high reliability and a high contrast, even by a curing reaction for a short time, whereby the state under application or non-application of an electric field, is stable. In Figure 1, a flow chart of one embodiment of the method for producing a liquid crystal optical element of the present invention, will be shown.

The curable sites (A_1 , A_2) of the formula (1) may be any of the above-mentioned functional groups which are photo curable or heat curable usually in the presence of a curing catalyst. Among them, an acryloyl group or a methacryloyl group suitable for photo curing, is preferred, since the temperature for the curing can be controlled.

The carbon numbers of the oxyalkylene portions R_1 and R_2 of the formula (1) are preferably from 2 to 6 from the viewpoint of the mobility. Further, a chain of an ethylene group having a carbon number of 2 and a propylene group having a carbon number of 3, are preferred.

As the mesogen structural portion (Z) of the formula (1), a bivalent polyphenylene having at least two 1,4-phenylene groups bonded, is preferred. Further, some of 1,4-phenylene groups in this polyphenylene group may be bivalent organic groups substituted by a 1,4-cyclohexylene group.

Some or all of the hydrogen atoms of such a polyphenylene group or a bivalent organic group may be substituted by a substituent such as a C_{1-2} alkyl group, a halogen atom, a carboxyl group or an alkoxycarbonyl group. Preferred Z is a biphenylene group having two 1,4-phenylene groups bonded (hereinafter referred to as a 4,4'-biphenylene group), a terphenylene group having three such phenylene groups bonded, and a bivalent organic groups having from 1 to 4 hydrogen atoms of such a group substituted by a C_{1-2} alkyl group, a fluorine atom, a chlorine atom or a carboxyl group. Most preferred Z is a 4,4'-biphenylene group having no substituent.

If n and m of the formula (1) are too large, the compatibility with the liquid crystal deteriorates, and each of them is from 1 to 10, further preferably from 1 to 4 taking into consideration the characteristics of the element after curing.

In the present invention, the uncured curable compound may contain two types of curable compounds, of which the molecular weights are different by at least two times, whereby the molecular weight between the crosslinking points of the resin during the curing process or after the curing may be changed. Or, the crystallinity of the resin may be controlled. The curing property in the curing process can be thus improved, and the modulus of elasticity of the resin after the curing

can be adjusted, whereby it is possible to obtain a liquid crystal optical element which is capable of presenting a high contrast even at a low driving voltage.

In order to adjust the compatibility with the liquid crystal before the curing and the modulus of elasticity of the resin after the curing, it is preferred that the uncured curable compound contains a curable compound containing a mesogen structural portion in its molecule and a curable compound containing no such a structural portion. Because, the mesogen structural portion improves the compatibility with the liquid crystal before the curing, while it tends to increase the modulus of elasticity of the resin after the curing more than necessary.

It is preferred that the contained two types of uncured curable compound can be bonded to each other, thereby to avoid phase separation of resins in the resin formed by curing and to avoid an increase of the haze at the time of reflection or transmission.

In order to lower the modulus of elasticity of the resin after curing by improving the curing property during the curing by increasing the molecular weight between the crosslinking points, it is preferred to employ a curable compound having a relatively large molecular weight as the uncured curable compound. Specifically, a curable compound having a molecular weight of at least 1000, is preferred.

The mixture of a liquid crystal with an uncured curable compound, may contain a curing catalyst, and in the case of photo curing, a photo polymerization initiator which is commonly used for a photo curable resin may be employed such as a benzoin ether type, an acetophenone type or a phosphine oxide type. In the case of thermosetting, a curing catalyst such as a peroxide type, a thiol type, an amine type or an acid anhydride type, may be used depending upon the type of the curable sites, and if necessary, a curing assistant such as an amine may also be used.

The content of the curing catalyst is preferably at most 20 wt% of the uncured curable compound contained, and in a case where a high molecular weight or a high resistivity is required for the cured product after curing, it is more preferably from 1 to 10 wt%.

Further, in order to improve the contrast of the element between application and non-application of an electric field, a chiral agent may be added to the mixture of a liquid crystal with an uncured curable compound. The helical pitch thereby induced is preferably at least 4 μm and at most three times of the electrode gap, since if it is too small, the driving voltage will increase, and if it is too large, no adequate contrast will be obtained.

If the helical pitch is smaller than 4 μm , there will be a problem that the transmittance at the time of

no application of a voltage tends to be low, or the driving voltage will increase. Further, if the helical pitch is larger than three times of the gap between the pair of electrodes sandwiching the liquid crystal/resin composite, the transmittance under application of a voltage will be high, and the contrast ratio in the transmittance between application and non-application of a voltage, tends to be low.

Further, by adjusting the helical pitch to be larger than 5 μm and at least two times of the electrode gap, it will be possible to adjust the balance of the low driving voltage and the high contrast.

In order to improve the compatibility with liquid crystal, the uncured curable compound in the mixture of liquid crystal with the curable compound, may contain a plurality of uncured curable compound differing in n and m in the formula (1), whereby the contrast may further be improved.

On the other hand, the mixture of a liquid crystal with an uncured curable compound, is preferably a homogeneous solution after mixing. Further, the mixture of a liquid crystal with an uncured curable compound may show a liquid crystal phase when sandwiched between the substrates provided with electrodes.

The mixture of a liquid crystal with an uncured curable compound, may show a liquid crystal phase when it is cured. It is also possible to impart a function to

align the liquid crystal to the electrode surface by directly rubbing the electrode surface of the substrates provided with electrodes, which sandwich the mixture of a liquid crystal with an uncured curable compound, or by forming a thin film of a resin thereof and rubbing the thin film, whereby it is possible to reduce irregularities at the time of sandwiching the mixture of a liquid crystal and an uncured curable compound.

Further, the combination of the alignment directions of the pair of alignment-treated substrates may be parallel or orthogonal, and the angle may be set to make the irregularities be minimum at the time of sandwiching the mixture.

The distance between the electrodes may be maintained by e.g. a spacer, and the gap is preferably from 5 to 50 μm , more preferably from 5 to 30 μm . If the electrode gap is too small, the contrast tends to deteriorate, and if it is too large, the driving voltage will increase. Figure 2 shows a schematic cross-sectional view of a liquid crystal optical element of the present invention.

It is a liquid crystal optical element 10 comprising glass substrates 1A and 1B, electrodes 2A and 2B, alignment films 3A and 3B and a liquid crystal/cured composite layer 4. It is an element which exhibits a transparent state at the time of non-application of a voltage and a scattering state at the time of application

of a voltage. Figure 3 is a view schematically illustrating a state in which a liquid crystal optical element of the present invention is used as a window glass of an automobile. It has a merit in that the transmittance in an oblique direction is high.

The substrates supporting electrodes, may be glass substrates or resin substrates, or a combination of a glass substrate and a resin substrate. Further, one side may be a reflecting electrode made of an aluminum or dielectric multi-layer film.

In the case of film substrates, the productivity is high, because it is possible that continuously supplied substrates provided with electrodes, are sandwiched between pairs of rubber rolls, and a mixture of a liquid crystal and an uncured curable compound, having a spacer incorporated and dispersed therein, is sandwiched between them, followed by continuous curing.

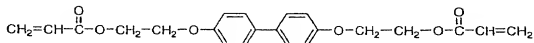
In the case of glass substrates, a very small amount of a spacer is distributed inside of the electrode surfaces, and the four sides of the opposing substrates are sealed with a sealing agent such as an epoxy resin to form a sealed cell, and one of cutouts of the seal formed at two or more portions is dipped in a mixture of a liquid crystal with an uncured curable compound, and suctioning from the other to fill the mixture into the cell, followed by curing to obtain a liquid crystal optical element. Otherwise, a vacuum injection method

may also be employed.

Now, the present invention will be described in detail with reference to Examples 1 to 7 (Examples of the present invention) and Examples A to E (Comparative Examples).

EXAMPLE 1

A mixture (mixture A) comprising 95 parts of a cyanotype nematic liquid crystal (BL-006, manufactured by Merck, dielectric anisotropy: positive), 5 parts of an uncured curable compound of the formula (4) and 0.15 part of benzoin isopropyl ether, was prepared.



Formula (4)

This compound of the formula (4) corresponds to a compound of the formula (1) wherein A_1 and A_2 are each an acryloyl group, R_1 and R_2 are each an ethylene group, the mesogen structural portion of Z is a 4,4'-biphenylene group, and each of n and m is 1.

This mixture A was injected into a liquid crystal cell prepared by disposing a pair of substrates having polyimide thin films formed on transparent electrodes and rubbed in one direction, to face so that the rubbing directions crossed each other, dispersing a very small amount of resin beads having a diameter of 13 μm , and bonding the substrates via the resin beads by an epoxy

resin printed along the four sides with a width of about 1 mm.

This liquid crystal cell was maintained at 25°C and ultraviolet rays of 3 mW/cm² from the upper side and
5 ultraviolet rays of about 3 mW/cm² from the lower side, were irradiated for 10 minutes by a HgXe lamp having a main wavelength of about 365 nm, to prepare a liquid crystal optical element.

An operation of applying a voltage of 50 Vrms with a
10 rectangular wave of 50 Hz to this liquid crystal optical element for 10 minutes and then removing the voltage, was repeated ten times. Then, the transmittance of the liquid crystal cell was measured by a transmittance measuring system (F value of the optical system: 11.5)
15 employing a measuring light source having a center wavelength of 530 nm and a full width at half maximum value of about 20 nm, whereby the transmittance was 79% in a state where no voltage was applied, 23% in a state where 50 Vrms was applied, and the difference in the
20 transmittance between application and non-application of the voltage, was 56%.

EXAMPLE A

A liquid crystal optical element was prepared in the same manner as in Example 1 except that an uncured
25 curable compound of the formula (3) (4,4'-bisacryloyloxybiphenyl) was used instead of the compound of the formula (4) as the uncured curable compound.

This compound of the formula (3) corresponds to a compound of the formula (1) wherein A_1 and A_2 are each an acryloyl group, the mesogen structural portion of Z is a 4,4'-biphenylene group, and each of n and m is 0.

5 To this liquid crystal optical element, a voltage was applied in the same manner as in Example 1. Then, the transmittance was measured by the same measuring system as in Example 1, whereby the transmittance was 72% in a state where no voltage was applied, 29% in a state
10 where 50 Vrms was applied, and the difference in the transmittance between the application and non-application of the voltage, was 43%.

EXAMPLE 2

A mixture (mixture B) having 2.5 wt% of a chiral
15 agent (a mixture of S-811, manufactured by Merck and C15 manufactured by Merck in a weight ratio of 1:1) dissolved in mixture A prepared in Example 1, was prepared.

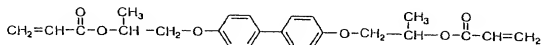
This mixture B was injected into the same liquid crystal cell as in Example 1, and while maintaining it at
20 25°C, ultraviolet rays of 3 mW/cm² from the upper side and ultraviolet rays of about 3 mW/cm² from the lower side were irradiated for 3 minutes by the same HgXe lamp having a main wavelength of about 365 nm as in Example 1, to obtain a liquid crystal optical element.

25 An operation of applying a voltage of 50 Vrms with a rectangular wave of 50 Hz to this liquid crystal optical element for 10 minutes and then removing the voltage, was

repeated ten times. Then, the transmittance was measured by a transmittance measuring system (F value of the optical system: 11.5) employing a measuring light source having a center wavelength of 530 nm and a full width at half maximum value of about 20 nm, whereby the transmittance was 78% in a state where no voltage was applied, and the value of a contrast obtained by dividing this value by the transmittance at the time when 50 Vrms was applied, was 33.

10 EXAMPLE 3

A liquid crystal optical element was obtained in the same manner as in Example 2 except that an uncured curable compound of the formula (5) was used instead of the compound of the formula (4) as the uncured curable compound.



Formula (5)

This compound of the formula (5) corresponds to a compound of the formula (1) wherein A₁ and A₂ are each an acryloyl group, R₁ and R₂ are each a propylene group, the mesogen structural portion of Z is a 4,4'-biphenylene group, and each of n and m is 1.

To this liquid crystal optical element, a voltage was applied in the same manner as in Example 2, and then the transmittance was measured by the same measuring

system, whereby the transmittance was 80% in a state where no voltage was applied, and the value of a contrast obtained by dividing this value by the transmittance at the time when 50 Vrms was applied, was 28.

5 EXAMPLE B

A liquid crystal optical element was obtained in the same manner as in Example 2 except that a compound of the formula (3) was used instead of the compound of the formula (4) as the uncured curable compound. To this
10 liquid crystal optical element, a voltage was applied in the same manner as in Example 2, and then the transmittance was measured by the same measuring system whereby the transmittance was 61% in a state where no voltage was applied, and the value of a contrast obtained
15 by dividing this value by the transmittance at the time when 50 Vrms was applied, was 17.

EXAMPLE 4

A mixture (mixture C) comprising 97 parts of one having 2.5 wt% of the chiral agent used in Example 2,
20 uniformly dissolved in a cyano type nematic liquid crystal (BL-009, manufactured by Merck), 3 parts of an uncured curable compound of the formula (4) and 0.09 part of benzoin isopropyl ether, was prepared.

This mixture C was injected into the same liquid
25 crystal cell as in Example 1, and while maintaining at 25°C, ultraviolet rays of 3 mW/cm² from the upper side and ultraviolet rays of about 3 mW/cm² from the lower

side, were irradiated for 30 minutes by the same HgXe lamp having a main wavelength of about 365 nm as in Example 1, to obtain a liquid crystal optical element.

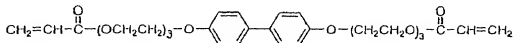
An operation of applying a voltage of 20 Vrms with a rectangular wave of 50 Hz to this liquid crystal optical element for 10 minutes and then removing the voltage, was repeated ten times. Then, the transmittance was measured by the same transmittance measuring system as mentioned above employing a measuring light source having a center wavelength of 530 nm and a full width at half maximum value of about 20 nm, whereby the transmittance was 82% in a state where no voltage was applied, and the value of a contrast obtained by dividing this value by the transmittance at the time when 20 Vrms was applied, was 11.

Further, an operation of applying a voltage of 30 Vrms with a rectangular wave of 50 Hz to this liquid crystal optical element for 10 minutes and then removing the voltage, was repeated ten times, and then the transmittance was measured in the same manner, whereby the transmittance was 82% in a state where no voltage was applied, and the value of a contrast obtained by dividing this value by the transmittance at the time when 30 Vrms was applied, was 40.

25 EXAMPLE 5

A mixture (mixture D) comprising 97 parts of one having 2.5 wt% of the chiral agent used in Example 2,

uniformly dissolved in a cyano type nematic liquid crystal (BL-009, manufactured by Merck), 2 parts of an uncured curable compound of the formula (4), 1 part of an uncured curable compound of the formula (6) and 0.09 part of benzoin isopropyl ether, was prepared.



Formula (6)

This compound of the formula (6) corresponds to a compound of the formula (1) wherein A_1 and A_2 are each an acryloyl group, R_1 and R_2 are each an ethylene group, the mesogen structural portion of Z is a 4,4'-biphenylene group, and each of n and m is 3.

This mixture D was injected into the same liquid crystal cell as in Example 1, and while maintaining it at 25°C, ultraviolet rays of 3 mW/cm² from the upper side and ultraviolet rays of about 3 mW/cm² from the lower side, were irradiated for 30 minutes by the same HgXe lamp having the main wavelength of about 365 nm as in Example 1, to prepare a liquid crystal optical element.

An operation of applying a voltage of 20 Vrms with a rectangular wave of 50 Hz to this liquid crystal optical element for 10 minutes and then removing the voltage, was repeated ten times. Then, the transmittance was measured by the same transmittance measuring system as mentioned above employing a measuring light source having a center

wavelength of 530 nm and a full width at half maximum value of about 20 nm, whereby the transmittance was 82% in a state where no voltage was applied, and the value of a contrast obtained by dividing this value by the transmittance at the time when 20 Vrms was applied, was 28.

EXAMPLE C

A liquid crystal optical element was obtained in the same manner as in Example 4 except that a compound of the formula (3) was used instead of the compound of the formula (4) as the uncured curable compound. An operation of applying a voltage of 20 Vrms with a rectangular wave of 50 Hz to this liquid crystal optical element for 10 minutes and then removing the voltage, was repeated ten times.

Then, the transmittance was measured by the same transmittance measuring system as mentioned above employing a measuring light source having a center wavelength of 530 nm and a full width at half maximum value of about 20 nm, whereby the transmittance was 57% in a state where no voltage was applied, and the value of a contrast obtained by dividing this value by the transmittance at the time when 20 Vrms was applied, was 10.

Further, an operation of applying a voltage of 30 Vrms with a rectangular wave of 50 Hz to this liquid crystal optical element for 10 minutes and then removing

the voltage, was repeated ten times, and then the transmittance was measured in the same manner as described above, whereby the transmittance was 49% in a state where no voltage was applied, and the value of a contrast obtained by dividing this value by the transmittance at the time when 30 Vrms was applied, was 6.

EXAMPLE 6

A mixture (mixture E) having 65 parts of a cyano type nematic liquid crystal (BL-006, manufactured by Merck), 35 parts of a chiral agent (a mixture of R-811, manufactured by Merck and CB15 manufactured by Merck in a weight ratio of 1:1), 3.1 parts of an uncured curable compound of the formula (4) and 0.09 part of benzoin isopropyl ether, was prepared.

This mixture E was injected into the same liquid crystal cell as in Example 1, and while maintaining it at 25°C, ultraviolet rays of 3 mW/cm² from the upper side and ultraviolet rays of about 3 mW/cm² from the lower side, were irradiated for 30 minutes by the same HgXe lamp having a main wavelength of about 365 nm as in Example 1, to prepare a liquid crystal optical element.

An operation of applying a voltage of 50 Vrms with a rectangular wave of 50 Hz to this liquid crystal optical element for 10 minutes and then removing the voltage, was repeated ten times. Then, the liquid crystal optical element was placed on black paper which did not

substantially reflect light, and the reflectance was measured by a reflectance measuring system (F value of the optical system: 8.2) employing a measuring light source having a center wavelength of 530 nm and a full width at half maximum value of about 20 nm, whereby the reflectance was 23% when no voltage was applied, 8% when 50 Vrms was applied, and the difference in reflectance between application and non-application of the voltage was 15%.

10 EXAMPLE D

A liquid crystal optical element was obtained in the same manner as in Example 6 except that a compound of the formula (3) was used instead of the compound of the formula (4) as the uncured curable compound. To this liquid crystal optical element, the voltage was applied in the same manner as in Example 5, and the reflectance was measured by the same measuring system, whereby the reflectance was 16% when no voltage was applied and 9% when 50 Vrms was applied, and the difference in reflectance between application and non-application of the voltage, was 7%.

EXAMPLE 7

A mixture (mixture F) comprising 95 parts of a nematic liquid crystal having a negative dielectric anisotropy ($T_c=98^\circ\text{C}$, $\Delta\epsilon=-5.6$, $\Delta n=0.220$), 5 parts of an uncured curable compound of the formula (4), and 0.15 part of benzoin isopropyl ether, was prepared.

This mixture F was injected into a liquid crystal cell prepared by bonding a pair of substrates having thin polyimide films for vertical alignment formed on transparent electrodes so that the thin polyimide films
5 faced each other, via a very small amount of resin beads of 6 μm , by an epoxy resin printed in a width of about 1 mm along the four sides.

While maintaining this cell at 25°C, ultraviolet rays of 3 mW/cm^2 from the upper side and ultraviolet rays of
10 about 3 mW/cm^2 from the lower side were irradiated for 10 minutes by a HgXe lamp having a main wavelength of about 365 nm, to obtain a liquid crystal optical element.

An operation of applying a voltage of 30 Vrms with a rectangular wave of 50 Hz to this liquid crystal optical
15 element for 10 minutes and then removing the voltage, was repeated ten times.

Then, the transmittance was measured by a transmittance measuring system (F value of the optical system: 11.5) employing a measuring light source having a
20 center wavelength of 530 nm and a full width at half maximum value of about 20 nm, whereby the transmittance was 86% in a state where no voltage was applied and 24% in a state where 50 Vrms was applied, and the difference in the transmittance between application and non-
25 application of the voltage, was 62%.

EXAMPLE E

A liquid crystal optical element was obtained in the same manner as in Example 7 except that a compound of the formula (3) was used instead of the compound of the formula (4) as the uncured curable compound. To this liquid crystal optical element, the voltage was applied in the same manner as in Example 7, and the transmittance was measured by the same measuring system, whereby the transmittance was 64% when no voltage was applied and 20% when 30 Vrms was applied, and the difference in transmittance between application and non-application of the voltage, was 44%. The results of the respective Examples are summarized and shown in the following Table 1.

Table 1

Example	Main materials used	conditions	T-von	T-voff	ΔT	CR
1	Formula (4)	After	79%	23%	56%	
2	Formula (4) + chiral agent		78%	About 2%		33
3	Formula (5) + chiral agent		80%	About 3%		28
4	Formula (4) + chiral agent		82%	About 7%		11
5	Formula (4) + Formula (6) + chiral agent	20 Vrms	82%	About 2%		40
6	Formula (4) + chiral agent	30 Vrms	82%	About 3%		28
7	Formula (4) + $\Delta \epsilon$ being negative	Reflection type	23%	8%	15%	
A	Formula (3)		86%	24%	62%	
B	Formula (3) + chiral agent		72%	29%	43%	17
C	Formula (3) + chiral agent	20 Vrms	61%	About 4%		
		30 Vrms	57%	About 6%		10
D	Formula (3) + chiral agent	Reflection type	49%	About 8%		6
E	Formula (3) + $\Delta \epsilon$ being negative		16%	9%	7%	
Note	CR represents the contrast ratio, and ΔT represents the differences between T-von and T-voff .					
			64%	20%	44%	

Now, with reference to Examples 8 to 11, the effects resulting from the fact that the uncured curable compound contains at least two types of curable compounds, of which the molecular weights are different by at least two times, will be described. Examples 8 and 10 represent cases where the effects are observed, and Examples 9 and 11 represent cases where no such effects are observed.

EXAMPLE 8

A mixture (mixture A) having 94.6 parts of a cyano type nematic liquid crystal (BL-009, manufactured by Merck), 2.4 parts of a chiral agent (a mixture of S-811, manufactured by Merck and C15 manufactured by Merck in a weight ratio of 1:1), 2.5 parts of a curable compound of the formula (4) having a molecular weight of 382, 0.5 part of an urethane acrylate oligomer (EB-270, manufactured by UCB) having a molecular weight of at least 1500 and 0.09 part of benzoin isopropyl ether, was prepared.

This mixture A was injected into a liquid crystal cell prepared by bonding a pair of substrates having thin polyimide films formed on transparent electrodes and rubbed in one direction, so that the rubbing directions crossed each other, via a very small amount of resin beads having a diameter of 13 μm , by an epoxy resin printed in a width of about 1 mm along the four sides.

While maintaining this liquid crystal cell at 25°C, ultraviolet rays of 3 mW/cm² from the upper side and

ultraviolet rays of about 3 mW/cm^2 from the lower side, were irradiated for 10 minutes by a HgXe lamp having a main wavelength of about 365 nm, to prepare a liquid crystal optical element.

5 An operation of applying a voltage of 20 Vrms with a rectangular wave of 50 Hz to this liquid crystal optical element for 10 minutes and then removing the voltage, was repeated ten times. Then, the transmittance was measured by a transmittance measuring system (F value of the
10 optical system: 11.5) employing a measuring light source having a center wavelength of 530 nm and a full width at half maximum value of about 20 nm, whereby the transmittance was 83% when no voltage was applied, and the value of a contrast ratio obtained by dividing this
15 value by the transmittance when 20 Vrms was applied, was 31.

EXAMPLE 9

A liquid crystal optical element was obtained in the same manner as in Example 8 except that only the compound
20 of the formula (4) was used as the uncured curable compound. To this liquid crystal optical element, the voltage was applied in the same manner as in Example 8, and the transmittance was measured by the same measuring system, whereby the transmittance was 83% when no voltage
25 was applied, and the value of a contrast ratio obtained by dividing this value by the transmittance when 20 Vrms was applied, was 11.

EXAMPLE 10

Mixture A prepared in Example 8 was injected into the same liquid crystal cell as in Example 8, and while maintaining it at 25°C, ultraviolet rays were irradiated
5 for 3 minutes in the same manner as in Example 8 to obtain a liquid crystal optical element.

An operation of applying a voltage of 20 Vrms with a rectangular wave of 50 Hz to this liquid crystal optical element for 10 minutes and then removing the voltage, was
10 repeated ten times. Then, the transmittance was measured by a transmittance measuring system (F value of the optical system: 11.5) employing a measuring light source having a center wavelength of 530 nm and a full width at half maximum value of about 20 nm, whereby the
15 transmittance was 79% when no voltage was applied, and the value of a contrast ratio obtained by dividing this value by the transmittance when 20 Vrms was applied, was 43.

EXAMPLE 11

20 A liquid crystal optical element was obtained in the same manner as in Example 10 except that only the compound of the formula (4) was used as the uncured curable compound.

To this liquid crystal optical element, the voltage
25 was applied in the same manner as in Example 10, and the transmittance was measured by the same measuring system, whereby the transmittance was 81% when no voltage was

applied, and the value of a contrast ratio obtained by dividing this value by the transmittance when 20 Vrms was applied, was 12.

Further, the effects resulting from the fact that a
5 chiral agent is incorporated to the above mixture, and the helical pitch of the chiral agent is adjusted to be at least 4 μm and at most three times of the electrode gap, will be described with reference to Examples 12 to 19. Examples 12, 13, 16 and 17 represent cases in which
10 such effects are observed, and Examples 14, 15, 18 and 19 represent cases where no such effects are observed.

EXAMPLE 12

A mixture having 3.5 parts of a chiral agent (a mixture comprising S-811, manufactured by Merck and C15
15 manufactured by Merck in a weight ratio of 1:1, hereinafter referred to as chiral agent A) uniformly dissolved in 100 parts of mixture A, was prepared (mixture H). Then, it was injected into a wedge cell for measuring the helical pitch, and the pitch was measured,
20 whereby the helical pitch was found to be 5.1 μm .

This mixture H was injected into a liquid crystal cell prepared by bonding a pair of substrates having thin polyimide films formed on transparent electrodes and rubbed in one direction, so that the rubbing directions
25 crossed each other, via a very small amount of resin beads having a diameter of 13 μm , by an epoxy resin printed in a width of about 1 mm along the four sides.

While maintaining this liquid crystal cell at 25°C, ultraviolet rays of 3 mW/cm² from the upper side and ultraviolet rays of about 3 mW/cm² from the lower side, were irradiated for 10 minutes by a HgXe lamp having a main wavelength of about 365 nm, to prepare a liquid crystal optical element.

An operation of applying a voltage of 50 Vrms with a rectangular wave of 50 Hz to this liquid crystal optical element for 10 minutes and then removing the voltage, was repeated ten times. Then, the transmittance was measured by a transmittance measuring system (F value of the optical system: 11.5) employing a measuring light source having a center wavelength of 530 nm and a full width at half maximum value of about 20 nm, whereby the transmittance was 78% when no voltage was applied, and the value of a contrast ratio obtained by dividing this value by the transmittance when 50 Vrms was applied, was 23.

In a case where the transmittance of this liquid crystal optical element when no voltage was applied, was 100%, and the transmittance when a voltage of 50 Vrms was applied, was 0%, the applied voltage showing a transmittance of 50% i.e. a value (V_{50}) of the applied voltage showing a change of 50% in transmittance, was 23 Vrms.

EXAMPLE 13

A mixture having 1.5 parts of the chiral agent A of

Example 12 uniformly dissolved in 100 parts of the mixture A of Example 12, was prepared (mixture I). In the same manner as in Example 12, it was injected into a wedge cell for measuring the helical pitch, and the pitch
5 was measured, whereby the helical pitch was 10.8 μm .

This mixture I was injected into a liquid crystal cell having the same structure as used in Example 12, and ultraviolet rays were irradiated in the same manner at 25°C to cure the uncured curable compound, to form a
10 liquid crystal optical element.

An operation of applying a voltage of 50 Vrms with a rectangular wave of 50 Hz to this liquid crystal optical element for 10 minutes and then removing the voltage, was repeated ten times. Then, the transmittance was measured
15 by the same transmittance measuring system as in Example 12, whereby the transmittance was 81% when no voltage was applied, and the value of a contrast ratio obtained by dividing this value by the transmittance when 50 Vrms was applied, was 23, and V_{50} was 19 Vrms.

20 EXAMPLE 14

Without adding any chiral agent, mixture A of Example 12 was injected into a liquid crystal cell having the same structure as used in Example 12, and ultraviolet rays were irradiated in the same manner at 25°C to cure
25 the uncured curable compound, to obtain a liquid crystal optical element.

In this liquid crystal cell, the alignment

directions were crossed each other, whereby as injected into the cell, mixture A shows a helical pitch of about four times of the distance between the electrodes of the cell, on appearance.

- 5 An operation of applying a voltage of 50 Vrms with a rectangular wave of 50 Hz to this liquid crystal optical element for 10 minutes and then removing the voltage, was repeated ten times. Then, the transmittance was measured by the same transmittance measuring system as in Example 10 12, whereby the transmittance was 79% when no voltage was applied, and the value of a contrast ratio obtained by dividing this value by the transmittance when 50 Vrms was applied, was 3.2, and V_{50} was 18 Vrms.

EXAMPLE 15

- 15 A mixture having 7.5 parts of the chiral agent A of Example 12 uniformly dissolved in 100 parts of the mixture A of Example 12, was prepared (mixture J). In the same manner as in Example 12, it was injected into a wedge cell for measuring the helical pitch, and the pitch 20 was measured, whereby the helical pitch was 2.4 μm .

- This mixture J was injected into a liquid crystal cell having the same structure as used in Example 12, and ultraviolet rays were irradiated in the same manner at 25°C, to cure the uncured curable compound, to form a 25 liquid crystal optical element.

An operation of applying a voltage of 50 Vrms with a rectangular wave of 50 Hz to this liquid crystal optical

element for 10 minutes and then removing the voltage, was repeated ten times. Then, the transmittance was measured by the same transmittance measuring system as in Example 12, whereby the transmittance was 73% when no voltage was applied, and the value of a contrast ratio obtained by dividing this value by the transmittance when 50 Vrms was applied, was 13, and V_{50} was 31.

EXAMPLE 16

Two parts of a chiral agent (a mixture comprising R-811, manufactured by Merck and CB15 manufactured by Merck in a weight ratio of 1:1, hereinafter referred to as chiral agent B) was uniformly dissolved in 100 parts of mixture A of Example 12 to prepare a mixture (mixture K). Then, it was injected into a wedge cell for measuring the helical pitch, and the pitch was measured, whereby the helical pitch was found to be 5.7 μm .

This mixture K was injected into a liquid crystal cell having the same structure as used in Example 12, and ultraviolet rays were irradiated for one minute in the same manner as in Example 12 at 25°C to cure the uncured curable compound, to form a liquid crystal optical element.

An operation of applying a voltage of 50 Vrms with a rectangular wave of 50 Hz to this liquid crystal optical element for 10 minutes and then removing the voltage, was repeated ten times. Then, the transmittance was measured by the same transmittance measuring system as in Example

12, whereby the transmittance was 81% when no voltage was applied, and the value of a contrast ratio obtained by dividing this value by the transmittance when 50 Vrms was applied, was 31, and V_{50} was 22 Vrms.

5 EXAMPLE 17

0.5 Parts of the chiral agent B of Example 16 was uniformly dissolved in 100 parts of the mixture A of Example 12 to prepare a mixture (mixture L). In the same manner as in Example 12, it was injected into a wedge
10 cell for measuring the helical pitch, and the pitch was measured, whereby the helical pitch was 21 μm .

This mixture L was injected into the same liquid crystal cell as used in Example 12, and ultraviolet rays were irradiated in the same manner as in Example 16 at
15 25°C, to cure the uncured curable compound, to obtain a liquid crystal optical element.

An operation of applying a voltage of 50 Vrms with a rectangular wave of 50 Hz to this liquid crystal optical element for 10 minutes and then removing the voltage, was
20 repeated ten times. Then, the transmittance was measured by the same transmittance measuring system as in Example 12, whereby the transmittance was 80% when no voltage was applied, and the value of a contrast ratio obtained by dividing this value by the transmittance when 50 Vrms was
25 applied, was 14, and V_{50} was 17 Vrms.

EXAMPLE 18

Without adding chiral agent, mixture A of Example 12

was injected into a liquid crystal cell having the same structure as used in Example 12, and ultraviolet rays were irradiated in the same manner as in Example 16 at 25°C, to cure the uncured curable compound, to obtain a
5 liquid crystal optical element.

This liquid crystal cell was a cell in which the alignment directions crossed each other, whereby as injected into the cell, mixture A shows a helical pitch of about 4 times of the distance between electrodes of
10 the cell, on appearance.

An operation of applying a voltage of 50 Vrms with a rectangular wave of 50 Hz to this liquid crystal optical element for 10 minutes and then removing the voltage, was repeated ten times. Then, the transmittance was measured
15 by the same transmittance measuring system as in Example 12, whereby the transmittance was 78% when no voltage was applied, and the value of a contrast ratio obtained by dividing this value by the transmittance when 50 Vrms was applied, was 3.9, and V_{50} was 10 Vrms.

20 EXAMPLE 19

Four parts of the chiral agent B of Example 16 was uniformly dissolved in 100 parts of the mixture A of Example 12 to prepare a mixture (mixture M). In the same manner as in Example 12, it was injected into a wedge
25 cell for measuring the helical pitch, and the pitch was measured, whereby the helical pitch was 3.0 μm .

This mixture M was injected into the same liquid

crystal cell as used in Example 12, and ultraviolet rays were irradiated in the same manner as in Example 16 at 25°C, to cure the uncured curable compound, to form a liquid crystal optical element.

- 5 An operation of applying a voltage of 50 Vrms with a rectangular wave of 50 Hz to this liquid crystal optical element for 10 minutes and then removing the voltage, was repeated ten times. Then, the transmittance was measured by the same transmittance measuring system as in Example 10 12, whereby the transmittance was 79% when no voltage was applied, and the value of a contrast ratio obtained by dividing this value by the transmittance when 50 Vrms was applied, was 25, and V_{50} was 28 Vrms.

INDUSTRIAL APPLICABILITY

- 15 The liquid crystal optical element of the present invention has high transmittance when it is transparent and has a high contrast or difference in transmittance between application and non-application of an electric field, and thus is suitable for e.g. an optical shutter 20 or a light-controlling glass which is required to provide a high light transmittance when it is transparent.

- Also when made into a reflection type liquid crystal optical element, it has a high reflectance at the time when no voltage is applied, and the contrast between 25 application and non-application of an electric field can be made high.

Further, the change in the voltage-transmittance

curve or the voltage-reflectance curve due to repetition of an operation of application and non-application of an electric field to the liquid crystal optical element, is small, whereby it is possible to present a highly
5 reliable liquid crystal optical element.

Especially when the above-mentioned curable compound contains two types of curable compounds, of which the molecular weights are different by at least two times, the curing property of the curable compound used will be
10 high, and it is possible to prepare a liquid crystal optical element having a high contrast in the transmittance or reflectance between application and non-application of an electric field, in a short period of curing time, whereby the productivity is high.

15 Further, the modulus of elasticity or the molecular weight between crosslinking points of the cured resin can be controlled, whereby a liquid crystal optical element showing a high contrast ratio, can be obtained even at a low driving voltage.

20 Further, when a chiral agent is incorporated to the mixture, and the helical pitch of the chiral agent is adjusted to be at least 4 μm and at most three times of the electrode gap, it is possible to obtain a liquid
25 crystal optical element which can be operated at a low driving voltage and which has a high contrast ratio in the transmittance between application and non-application of an electric field.

Further, the transmittance of the element when it is transparent, can be made high, and irregularities in the transmittance derived from the injection step or the sandwiching step can be made small, whereby it is possible to provide a liquid crystal optical element suitable for e.g. a high quality light-controlling glass or optical shutter.

Further, the contrast ratio can be substantially improved at a low voltage without substantially increasing the driving voltage, whereby it can be used for a display element.

CLAIMS

1. A method for producing a liquid crystal optical element, which comprises sandwiching a mixture of a liquid crystal with an uncured curable compound between a pair of substrates which are provided with transparent electrodes and of which at least one is transparent, and curing the curable compound to form a liquid crystal/cured composite layer, wherein the curable compound contains a compound of the formula (1):
- 10 $A_1-(OR_1)_n-O-Z-O-(R_2O)_m-A_2$ Formula (1)
- wherein each of A_1 and A_2 which are independent of each other, is an acryloyl group, a methacryloyl group, a glycidyl group or an allyl group; each of R_1 and R_2 which are independent of each other, is a C_{2-6} alkylene group; Z
- 15 is a bivalent mesogen structure; and each of n and m which are independent of each other, is an integer of from 1 to 10.
2. The method for producing a liquid crystal optical element according to Claim 1, wherein Z is a 4,4'-
- 20 biphenylene group, or a 4,4'-biphenylene group having part or all of hydrogen substituted by C_{1-2} alkyl or halogen atoms.
3. The method for producing a liquid crystal optical element according to Claim 1 or 2, wherein each of R_1 and
- 25 R_2 which are independent of each other, is an ethylene group or a propylene group.
4. The method for producing a liquid crystal optical

element according to Claim 1, 2 or 3, wherein each of A_1 and A_2 which are independent of each other, is an acryloyl group or a methacryloyl group.

5. The method for producing a liquid crystal optical
5 element according to Claim 1, 2, 3 or 4, wherein each of n and m which are independent of each other, is from 1 to 4.

6. The method for producing a liquid crystal optical
10 element, wherein the curable compound contains two types of curable compounds, of which the molecular weights are different by at least two times.

7. The method for producing a liquid crystal optical
15 element according to Claim 6, wherein the curable compound contains a curable compound containing a mesogen structural portion in its molecule and a curable compound containing no mesogen structural portion.

8. The method for producing a liquid crystal optical
20 element according to Claim 6 or 7, wherein the two types of curable compounds have curable sites connectable to each other.

9. The method for producing a liquid crystal optical
element according to Claim 6, 7 or 8, which contains a curable compound having a molecular weight of at least 1,000.

25 10. The method for producing a liquid crystal optical element according to any one of Claims 1 to 9, wherein the mixture contains a chiral agent.

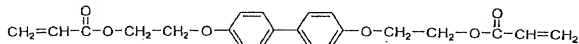
11. The method for producing a liquid crystal optical element according to any one of Claims 1 to 9, wherein the mixture contains a chiral agent, and the helical pitch of the chiral agent is at least 4 μm and at most three times of the electrode gap.
12. The method for producing a liquid crystal optical element according to Claim 11, wherein the electrode gap is from 4 to 50 μm .
13. The method for producing a liquid crystal optical element according to Claim 11 or 12, wherein the helical pitch is at least 5 μm and at most two times of the electrode gap.
14. The method for producing a liquid crystal optical element according to any one of Claims 1 to 13, wherein the mixture contains a very small amount of a curing catalyst.
15. The method for producing a liquid crystal optical element according to any one of Claims 1 to 14, wherein a plurality of compounds of the formula (1) wherein n and m are different, are used in combination.
16. A liquid crystal optical element produced by the method as defined in any one of Claims 1 to 15.

ABSTRACT

A liquid crystal/cured composite layer is formed by sandwiching a mixture of a liquid crystal with an uncured curable compound i.e. a curable compound represented by a
5 structure of an acryloyl group- $(OR_1)_n$ -O-mesogen structural portion-O- $(R_2O)_m$ -acryloyl group structure (wherein each of R_1 and R_2 is a C_{2-6} alkylene group, and each of n and m is from 1 to 10), between a pair of
10 substrates provided with transparent electrodes, and curing the curable compound.

Fig. 1

A MIXTURE OF A COMPOUND OF THE FORMULA:



WITH A LIQUID CRYSTAL, IS SANDWICHED BETWEEN A PAIR OF SUBSTRATES PROVIDED WITH ELECTRODES



THE CURABLE COMPOUND IS CURED

Fig. 2

1 0
✓

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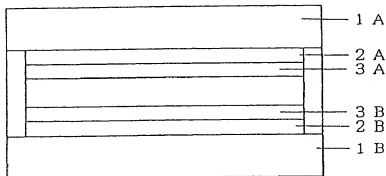
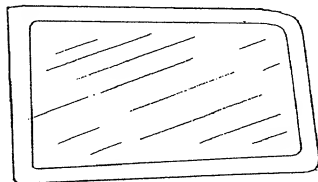


Fig. 3



Declaration and Power of Attorney For Patent Application

特許出願宣言書及び委任状

Japanese Language Declaration

日本語宣言書

下記の氏名の発明者として、私は以下の通り宣言します。

As a below named inventor, I hereby declare that:

私の住所、私書箱、国籍は下記の私の氏名の後に記載された通りです。

My residence, post office address and citizenship are as stated next to my name.

下記の名称の発明に関して請求範囲に記載され、特許出願している発明内容について、私が最初かつ唯一の発明者（下記の氏名が一つの場合）もしくは最初かつ共同発明者（下記の名称が複数の場合）であると信じています。

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled.

LIQUID CRYSTAL OPTICAL ELEMENT

AND METHOD FOR ITS PRODUCTION

上記発明の明細書は、

the specification of which

☐ 本書に添付されています。

☐ is attached hereto.

☒ 月 日 に提出され、米国出願番号または特許協定条約国際出願番号を _____ とし、
(該当する場合) _____ に訂正されました。

☒ was filed on October 19, 1999
as United States Application Number or
PCT International Application Number
PCT/JP99/05765 and was amended on
_____ (if applicable).

私は、特許請求範囲を含む上記訂正後の明細書を検討し、内容を理解していることをここに表明します。

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

私は、連邦規則法典第37編第1条56項に定義されるとおり、特許資格の有無について重要な情報を開示する義務があることを認めます。

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56.

Japanese Language Declaration
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私は、米国法典第35編119条 (a) - (d) 項又は365条 (b) 項に基づき下記の、米国以外の国の少なくとも一カ国を指定している特許協力条約365 (a) 項に基づく国際出願、又は外国での特許出願もしくは発明者証の出願についての外国優先権をここに主張するとともに、優先権を主張している、本出願の前に出願された特許または発明者証の外国出願を以下に、枠内をマークすることで、示しています。

Prior Foreign Application(s)

外国での先行出願

10-298620

Japan

(Number)
(番号)

(Country)
(国名)

10-298621

Japan

(Number)
(番号)

(Country)
(国名)

10-298624

Japan

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(Application No.)
(出願番号)

(Filing Date)
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I hereby claim foreign priority under Title 35, United States Code, Section 119 (a)-(d) or 365(b) of any foreign application(s) for patent or inventor's certificate, or Section 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

Priority Claimed

優先権主張

<input checked="" type="checkbox"/> Yes	<input type="checkbox"/> No
はい	いいえ
<input checked="" type="checkbox"/> Yes	<input type="checkbox"/> No
はい	いいえ
<u>Yes</u>	

20/October/1998

(Day/Month/Year Filed)
(出願年月日)

20/October/1998

(Day/Month/Year Filed)
(出願年月日)

20/October/1998

I hereby claim the benefit under Title 35, United States Code, Section 119(e) of any United States provisional application(s) listed below.

(Application No.)
(出願番号)

(Filing Date)
(出願日)

I hereby claim the benefit under Title 35, United States Code, Section 120 of any United States application(s), or Section 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of Title 35, United States Code Section 112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of application.

(Status: Patented, Pending, Abandoned)
(現況: 特許許可済、係属中、放棄済)

(Status: Patented, Pending, Abandoned)
(現況: 特許許可済、係属中、放棄済)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Japanese Language Declaration
(日本語宣言書)

委任状：私は下記の発明者として、本出願に関する一切の手続きを米特許商標局に対して遂行する弁理士または代理人として、下記の者を指名いたします。
(弁護士、または代理人の指名及び登録番号を明記のこと)

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith: (list name and registration number)

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